An experiment study on Raman spectrum of liquid core optical Fiber
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ABSTRACT
The Raman spectra intensity can be enhanced in liquid core optical fiber (LCOF). The total Raman power emitted by the LCOF is a function of the following parameters: LCOF length, LCOF loss coefficients at the laser and Raman wavelengths, the concentration of analyte and input laser power. The attenuation of the light in CS₂ is small in the range from 1400nm to 1700nm. We dissolved liquid CCL₄ into CS₂ and got different solutions of different concentrations. When the analyte concentration was changed, the analyte molecules and the numerical aperture of the LCOF were also changed, so there is an optimum concentration at which the maximum Raman spectrum can be obtained. Our experiment result is in good agreement with the calculated results. Backward Raman scattering is superior to forward Raman scattering in our experiment. Raman intensity first increases to a maximum with fiber length, then decrease because the fiber attenuation becomes dominant. Therefore, there exists the optimum fiber length. In our experiment, we find that higher Raman intensity can be obtained when the fiber length is 2m. The Raman intensity becomes powerful when the input pump power becomes larger. Raman linewidth becomes narrow when the concentration decreased. Our experiment also approves it.

Keywords: Liquid core optical fiber; Raman spectrum; Raman linewidth

1. INTRODUCTION
C.V.Raman found Raman scattering phenomenon in 1928. After that, Raman spectrum technology has become an important part of spectroscopy. Since Liquid core optical fiber (LCOF) of benzene core was made in American Bell Laboratory in 1970s, LCOF was used widely in many science areas. The reasons of studying on Raman spectrum in LCOF is that laser can be transmitted in LCOF, then it can contact with the sample sufficiently. Since the fiber may be many meters long, much more Raman light can be produced and collected than would be conceivable with conventional systems. Raman intensity can be improved. 1-3
The total Raman power emitted by the LCOF is a function of the following parameters: LCOF length, LCOF loss coefficients at the laser and Raman wavelengths, the concentration of analyte and input laser power. Many areas most likely to benefit from the use of LCOFs include the following: (1)environment monitoring, which necessitates the ability to detect low-level analyte concentrations in aqueous solutions; (2)low the cost the device, where the intensity enhancement provided by LCOFs might enable the use of less expensive lasers and detectors; (3)high-performance liquid chromatography, where convenient geometry, small volume, and improved signal intensity and signal-to-noise ratio combine to make Raman detection attractive; (4)biological applications, where low solubility and denaturing at high laser power often mandate the use of nonstandard sampling techniques4. Therefore, it is important to study on Raman spectrum of liquid core optical fiber.

2. EXPERIMENTAL SETUP
Figure 1 is the measuring experimental set-up of Raman spectrum of liquid-core optical fiber. Backward Raman scattering is measured. There are several sections as following below:

1. Liquid core optical fiber (LCOF)
   The internal diameter of hollow core silica optical fiber is 150um, and it is tenderness and can not be easy to break off because of the protecting layer covered in the fiber surface. It is filled with different concentration of the solution which is made of CS$_2$ and CCl$_4$, and ended with a container filled with the solution.

2. Single mode fiber (SM)
   The internal diameter of single mode fiber is 9um and the external diameter is 125um. Since the external diameter of SM is smaller than the internal diameter of LCOF, SM can be put into LCOF directly.

3. Wavelength division multiplexer
   1427/1520 CWDM is adopted. Pump source is fiber Raman laser. Wavelength is 1427.2nm. Bandwidth is 0.067nm. Tunable range of output power is 1 to 1200mw.

4. Optical Spectrum Analyzer (OSA)
   Spectrum range: 600nm-1700nm; Resolution: 10pm; Dynamic range: 60dB

3. RESULTS AND DISCUSSION

3.1 The core of the fiber
The intensity of CCl$_4$ ($n_{CCl_4}=1.457$) Raman spectrum is much larger among kinds of liquid. Unfortunately, the refractive index is smaller than that of quartz ($n_{quartz}=1.460$). Therefore, the enhanced optical fiber Raman spectrum can not be acquired directly. So we must choose a solvent of which refractive index is larger than quartz such as C$_6$H$_6$, C$_7$H$_8$, C$_3$H$_8$O, CS$_2$. Attenuation spectrum of the liquids mentioned above were measured and shown in fig2. The attenuation of the light in CS$_2$ is small in the range between 1400nm to 1700nm. We dissolved liquid CCl$_4$ into CS$_2$ and got different solution of different concentration. We measured the refractive index with a series of different CCl$_4$ concentration from 0 to 1 and obtained a linear relation. Fig2 shows the fitting.
between the experimental data of the refractive index and the formula is

\[ n_{\text{core}} = cx + d \]  

(1)

where \( c = -0.157, d = 1.60936 \) for solvent CS\(_2\) and \( x \) is analyte concentration. The use of different concentration results in different refractive indexes of the liquid core optical fiber. Hence, the exit numerical aperture will decrease with a lower analyte concentration, resulting in a decreased Raman signal, the light throughput being related to the square of the numerical aperture \( A \).

### 3.2 Raman intensity influenced by the concentration

While Raman scattering occurs in all directions, only the portion within the acceptance angle undergoes total internal reflection and travels down the core. The acceptance angle \( \theta_a \) shown in figure 3 is given by

\[ \theta_a = 90^\circ - \theta_c = 90^\circ - \arcsin \left( \frac{NA}{n_{\text{core}}} \right) \]  

(2)

where \( \theta_c \) is the critical angle for total internal reflection, where \( n_{\text{core}} \) and \( n_{\text{clad}} \) are the refraction indexes of the liquid and quartz, respectively.

Numerical aperture (NA) represents the ability of receiving light in fiber.

\[ NA = \sqrt{(n_{\text{core}})^2 - (n_{\text{clad}})^2} \]  

(3)

When the value of NA is bigger, the ability of receiving light is more powerful and versa versa. The exist numerical aperture of the liquid core optical fiber will change with the analyte concentration. The Raman intensity relates to the two factors mentioned above and it also can be influenced by the number of analyte.
molecules which is in direct proportion to the analyte concentration. Therefore, 
\[ I = \theta_0 \cdot N_A \cdot x \cdot N_A^2 \cdot x \]  
(4)

\( n_{\text{clad}} \) is equal to \( n_{\text{quartz}} \).

From (1)(2)(3)(4), we get formula (5) which was shown below.

\[ I = 0.025x^3 - 0.505x^2 + 0.458x \]  
(5)

Therefore, the optimal concentration that was calculated from the above formula is 0.46. Fig 4 shows the comparison between the measurement and the calculation of normalized Raman intensity. We choose the intensity of 459 cm\(^{-1}\) Raman peak of CCl\(_4\) when the pump power is 0.5w as the measurement value referred in figure 4. The calculated value in figure 4 results in excellent agreement with the experiment. So, the three factors can reflect Raman intensity of liquid core optical fiber.

![Figure 4 the comparison between the measurement and the calculation of normalized intensity](image)

In the proceeding of experiment, the stability is worse along with the pump power become powerful when the analyte concentration is greater. As long as the pump power is more than 0.5w, the noise detected is larger and SN figure decreased. However, when the concentration is smaller, higher pump power can be put into the fiber and stronger Raman peak can be obtained. The phenomenon occurred because the following reasons: (1) when high pump power is coupled into the fiber, the solution tends to be gasified because of higher content of CCl\(_4\). (2) When the analyte concentration is higher, the numerical aperture is lower, then the ability of receiving light in fiber is bad, and it is difficult to detect the Raman light.

3.3 Raman intensity influenced by the length of LCOF

Backward Raman scattering is superior to forward Raman scattering in our experiment. If stimulated effects are neglected, the intensification of the Raman scattered light \( dI_R \) in the fiber in length \( dx \) is given by

\[ dI_R (x) = G_R I(x) dx - \alpha dI_R (x) dx \]  
(6)

\( G_R \) is Raman gain coefficient which is related to Raman scattering cross section, \( I_R \) and \( I \) are the Raman and exciting intensities, and \( \alpha \) is the loss coefficient for light traveling in the fiber. If it is assumed for simplicity that the loss coefficient in the fiber is the same at the wavelength of the Raman or exciting radiations, and that the amount of light converted to Raman radiation is small, then

\[ I(x) = I_0 e^{-\alpha x} \]  
(7)

where \( I_0 \) is the intensity of the exciting light at \( x=0 \). Substitute Eq. (7) into Eq. (6) gives

\[ I_R = \int dI_R (x) = G_R I_0 x e^{-\alpha x} \]  
(8)
From (8), we can conclude that Raman intensity first increases to a maximum with fiber length, then decreases because the fiber attenuation becomes dominant. Therefore, there exist the optimum fiber length.

\[
\frac{dI_R}{dx} = 0 = -G_R I_0 \alpha e^{-\alpha x} + G_R I_R e^{-\alpha x}
\]  

(9)

Therefore,

\[
L = \frac{1}{\alpha}
\]

(10)

Thus, the optimum length L is 1/\( \alpha \).

In our experiment, we measured Raman intensity of 459\( \text{cm}^{-1} \) Raman shift along with the changed fiber length and shown in Table 1. When the fiber length is 2m, the Raman intensity is larger. Meanwhile, we can see that the Raman intensity becomes powerful when the pump power becomes larger.

Table 1 Raman intensity changed with the fiber length and pump power

<table>
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<tr>
<th>Fiber length</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
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<tr>
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<td>0.72</td>
<td>0.71</td>
<td>0.71</td>
</tr>
</tbody>
</table>

3.4 Raman linewidth influenced by the concentration

The Raman linewidth of organic solution CCl\(_4\) (8\( \text{cm}^{-1} \)) is narrow compared with silica (several hundred wavenumbers) [5]. It is reported that Raman linewidth becomes narrow when the concentration decreased. Our experiment also approve it and shown in Fig 5.

![Figure 5. Raman linewidth changed with different concentration](image)

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